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14. ABSTRACT
This research explored the potential for backward lasing from natural constituents in air with the objective of developing revolutionary methods for standoff detection of trace gases. Backward lasing enables far-field monitoring through modulation methods. The approach utilizes the very high gain available from multiphoton pumping of atomic species, which can be created by the dissociation of molecules in air. Lasing has been demonstrated from oxygen, nitrogen and water vapor. In each of these cases the lasing is achieved by two photon pumping of the atomic fragments, leading to high gain backward lasing from oxygen, nitrogen, and hydrogen atoms. The research has demonstrated that predissociation significantly enhances the efficiency, and in the cases of nitrogen and water vapor it is essential. Backward lasing from two simultaneously pumped, closely separated regions in the air provides a method for the reduction of pulse to pulse fluctuations. Further reduction in pulse to pulse fluctuations will be possible if the dissociate step is eliminated by using argon, which requires three photon pumping. Backward lasing from argon in air has been achieved, but at argon concentrations of 10%. Future work is expected to reduce this threshold to the ~1% natural concentration of argon in air.

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Air Standoff Detection of Trace Molecules by Remote High Gain Backward Lasing in Air

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Abstract

This research explored the potential for backward lasing from natural constituents in air with the objective of developing revolutionary methods for standoff detection of trace gases. Backward lasing enables far-field monitoring through modulation methods. The approach utilizes the very high gain available from multiphoton pumping of atomic species, which can be created by the dissociation of molecules in air. Lasing has been demonstrated from oxygen, nitrogen and water vapor. In each of these cases the lasing is achieved by two photon pumping of the atomic fragments, leading to high gain backward lasing from oxygen, nitrogen, and hydrogen atoms. The research has demonstrated that predissociation significantly enhances the efficiency, and in the cases of nitrogen and water vapor it is essential. Backward lasing from two simultaneously pumped, closely separated regions in the air provides a method for the reduction of pulse to pulse fluctuations. Further reduction in pulse to pulse fluctuations will be possible if the dissociate step is eliminated by using argon, which requires three photon pumping. Backward lasing from argon in air has been achieved, but at argon concentrations of 10%. Future work is expected to reduce this threshold to the ~1% natural concentration of argon in air.

OBJECTIVES

The objectives of this research program have been to establish high sensitivity stand off detection methodologies based on backward lasing from natural species in air. Once such a method has been developed it can be used for trace detection by utilizing it in conjunction with a modulating laser that is tuned to the specific trace species of interest, causing a modulation either to the outward propagating pump laser or to the backward propagating air laser. That modulation is detected relative to a similar non modulated backward propagating air laser beam. The technical approach is to use a short pulse UV pump laser to produce a remote population inversion in an atomic species, leading to “cavityless” lasing. Lasing occurs from the population inversion that is created in the focal volume of the pump laser, and the lasing direction is determined by the geometry of that volume, reflecting the exponential amplification of stimulated emission with path length. Due to the multiphoton nature of the pumping, the gain volume is well localized to the high intensity region of the pump. That gain volume is elongated in the propagation direction of the pump laser and localized to the Rayleigh range of the laser focal volume. This leads to lasing in the forward and backward directions, with the backward

lasing beam retracing the pump path in the backward direction and the forward lasing overlapping the continued propagation of the pump beam in the forward direction. The atomic species are created by dissociation of oxygen, nitrogen or water vapor either with the same pulse that pumps the atomic fragments, or with an earlier pulse focused to overlap the same volume.

Figure 1 shows the atomic energy levels associated with the hydrogen, nitrogen and oxygen air lasers. Hydrogen is two photon pumped with 205nm light, nitrogen with 207 nm or 211 nm light and oxygen with 226 nm light. Lasing can be achieved with picosecond and nanosecond pumping, and, possibly, with femtosecond pumping. The general configuration for the experiments is shown in figure 2, indicating that both backward and forward lasing are monitored as well as emission to the side. Figure 3 shows the approximate geometry of the gain region, which is formed by the focal zone of the pump laser. Since the pump in all cases is in the ultraviolet and the lasing is in the red or infrared, separation of the pump from the lasing is easily accomplished with dichroic mirrors or prisms.

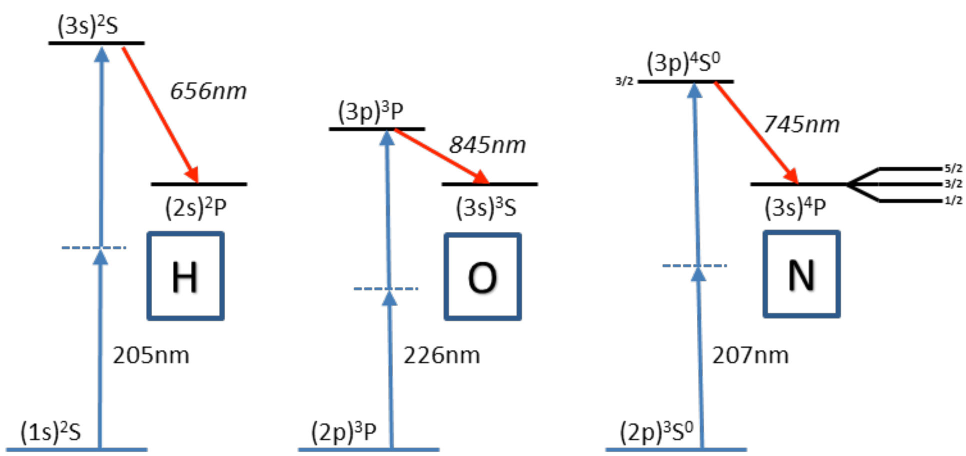

Figure 1: Energy levels for 2-photon pumping in hydrogen, oxygen, and nitrogen atoms.

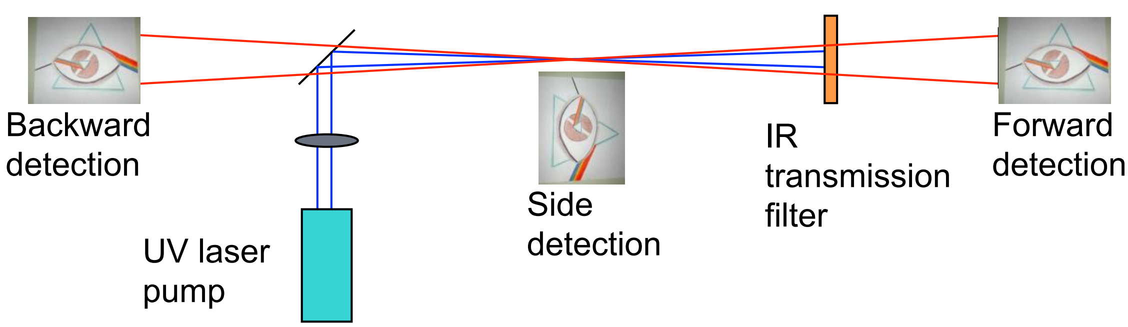

Figure 2: Experimental setup for forward and backwards air lasing.

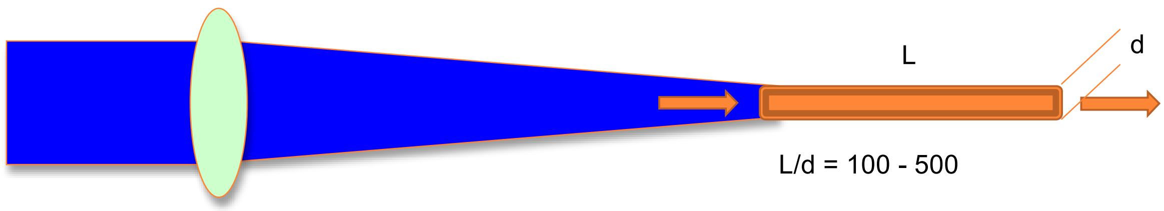

Figure 3: Mechanism for air lasing: The geometry of the gain region allows for high gain along the propagation direction of the pump beam

Both oxygen lasing and nitrogen lasing arise from major species in the air, however hydrogen lasing occurs from water vapor at as low as 40% relative humidity in room air, corresponding to a mole fraction below 1%. Figure 4 (right) shows the relative backward hydrogen lasing pulse energy as a function of humidity in 25C room air. When pumped by a 100 psec laser, a preliminary dissociation step is required. For these experiments, the water molecule was dissociated using either a 10 nanosecond Nd:YAG laser operating with 200 mJ at 1064nm or a 50 fsec Ti:sapphire laser operating with 1 mJ at 800 nm. It is interesting to note that the hydrogen laser pulse arises from the hydrogen Balmer alpha line (see Figure 1, left), which is at 656.3 nm (Figure 4, left), essentially the same color as a red laser pointer.

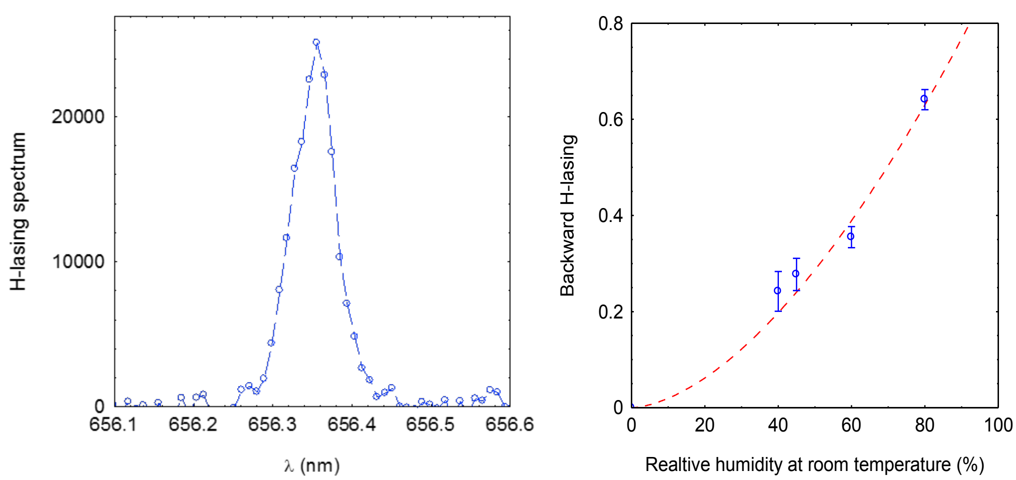

Figure 4. Left: Hydrogen backwards lasing spectrum. Right: Hydrogen lasing in air as a function of the humidity shows nonlinear dependence of the emission on the concentration of water molecules.

In order to use the backward lasing for the detection of trace species, the outward propagating laser beam will be modulated. There is an intrinsic pulse-to-pulse variation in the backward lasing due to the highly nonlinear nature of the dissociation and pumping processes as well as the variations in propagation through the air. In order to provide a reference that is capable of removing these pulse-to-pulse variations, a second, simultaneous backward lasing beam is generated using the same UV pumping pulse, split with a beam splitter and focused to a stand off location very close to the first backward lasing location. In this manner the pulse-to-pulse fluctuations of the second backward lasing beam are almost identical to those of the first, providing a pulse-to-pulse reference. With this arrangement modulation of the first backward lasing beam for tract detection can be referenced to the

second beam. Figure 5 shows this configuration, with the "second laser" providing the modulation. Figure 6 shows preliminary results from this dual beam configuration. The red squares and blue circles indicate the return pulse energy of the two beams for a series of 1000 laser pulses. The variations are up to 72% of the mean, and in some cases the return pulse energy approached zero. The green filled in circles show the ratio of the two pulses, computed on a pulse-to-pulse basis. The fluctuations are reduced to 1.7% of the mean.

The residual fluctuations are most likely due to the dissociation step, which can be highly variable. Efforts are underway to determine if natural atomic species in air such as argon or krypton can be used for backward lasing. Argon has a molar concentration of 0.8% in air, approximately the same as the water vapor, however it requires a three-photon pumping step rather than the two-photon steps available for oxygen, nitrogen and hydrogen. Figure 7 (left) shows the three-photon pumping of argon at 261 nm leading to backward lasing at 1327 nm. Figure 7 (right) shows the relative pulse energy of the backward lasing as a function of argon concentration in pure argon and in air. Note that in pure argon the backward lasing remains robust to below 10% of atmospheric density, however in air the signal is lost at 10%, somewhat more than a factor of ten higher than the natural density of argon in air. Work in the follow on research effort is addressing this limitation using a higher pulse energy laser source.

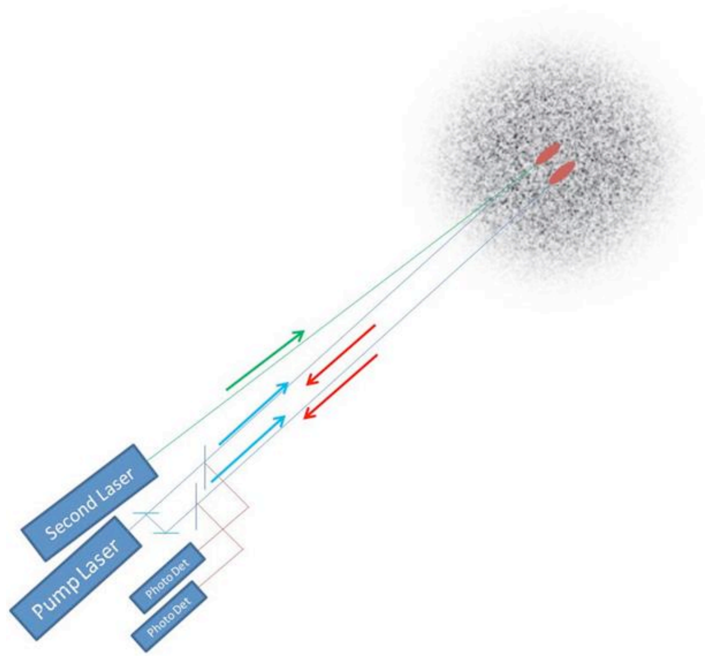

Figure 5: Dual backward lasing configuration for simultaneous reference backward beam generation providing enhanced detectability of the modulation of the backward lasing process. The "second laser" is tuned to a trace species and causes a modulation of one of the backward lasing beams relative to the other.

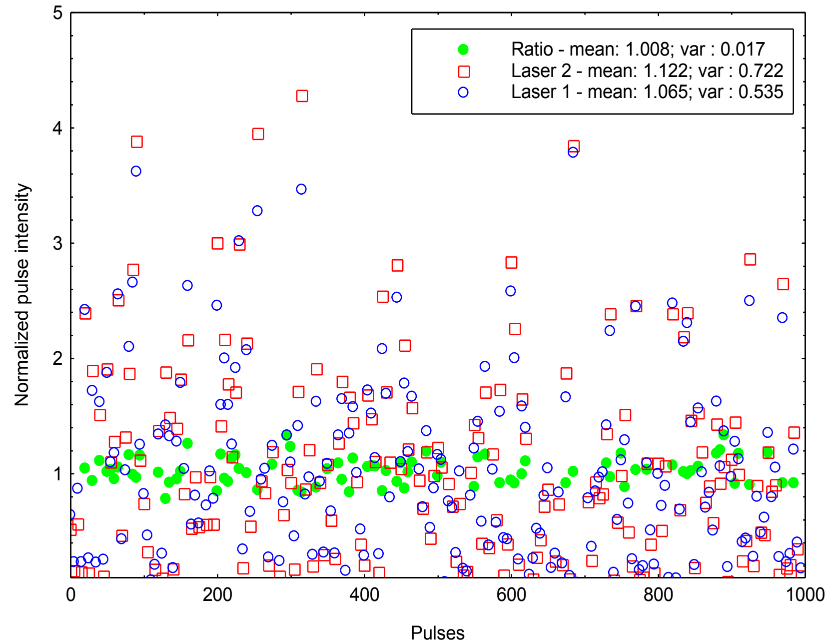

Figure 6: The reduction of pule to pulse variability using the ratio of the two simultaneous backward lasing pulses. The pulse to pulse variability of the individual backward lasing is up to 72% of the mean. The variability of the ratio of the two is less than 2%.

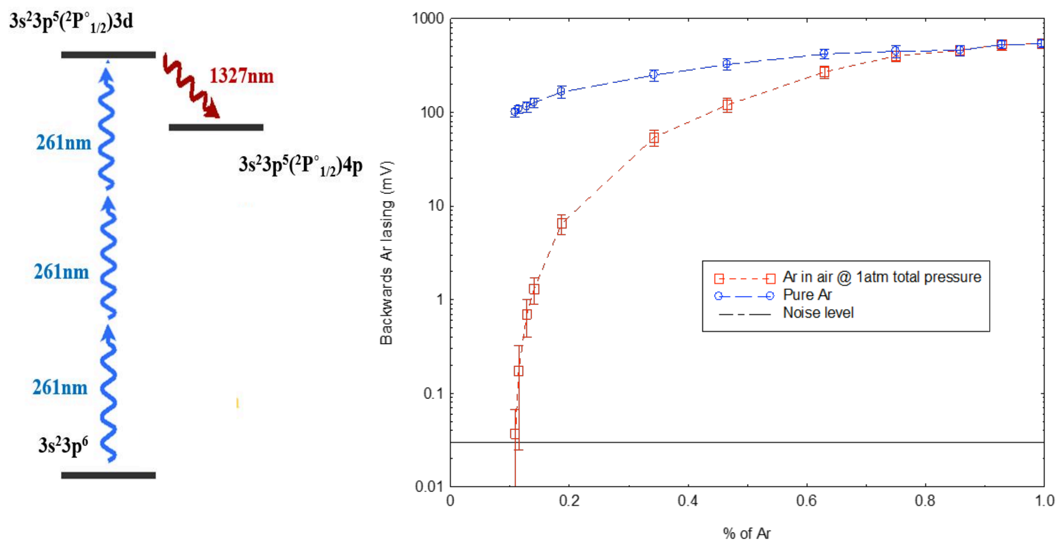

Figure 7: Backward lasing from argon in air. Left: the three photon pumping at 261 nm leads to lasing at 1327 nm. Right: The backward lasing from reduced concntrations of argon, in pure argon (blue) and in atmospheric pressure air (red).

WORK COMPLETED

High gain backward laser beams from two photon pumped atomic oxygen, nitrogen and hydrogen produced by dissociation of molecules in room air all posess similar characteristics: high spatial and temporal coherence and close to transform limited properties with pulse lengths of between 10 and 30 psec, independent of the pulse length of the pumping laser. The laser beams are generated over millimeter scale paths and reflect exponential gains corresponding to approximately $e^{60/cm}$. The exponential dependence of the gain on path length and atom number density and the lack of sensitivity to dephasing collision rates all indicate that the mechanism for the generation of the backward lasing is stimulated emission. Backward lasing from two simulatenously pumped regions provides a pulse to pulse reference for potential trace species dtection methods based on the modulation of the backward lasing. Residual variability arising from the dissociation step suggests that backward lasing from naturally present atomic species such as argon and krypton would be beneficial. Preliminary work indicates that three photon pumped backward lasing from argon might be feasible. Backward lasing from argon at concentrations as low as 10% in air has been achieved.

RESULTS

Demonstration of high gain lasing from two photon pumping of oxygen, nitrogen and hydrogen atoms following dissociation of molecular oxygen and nitrogen. Demonstration for the first time lasing in air from a minor species in air: atomic hydrogen obtained by dissociating the water molecules. This strong emission at 656nm is greatly enhanced by pre-dissociation with a non-resonant laser pulse, and it can be used to induce the stimulated emission necessary to optically pump the H atoms into a state with m-number selectivity. Preliminary results have been achieved on backward lasing from argon in air at mole fractions down to 10%.

IMPACT/APPLICATIONS

The research has established the potential for backward lasing in air from numerous atomic species. Backward lasing may provide a high sensitivity method for the detection of greenhouse gases, gas leakage from pipelines and refineries, pollution, illicit chemical and nuclear processing activities, chemical gas attacks, and the presence of explosives and hazardous materials. Other applications of high gain air lasing are of significant interest and include "around the corner" illumination, clandestine communication, and a local "guide star" for the correction of aero-optical distortion.

RELATED PROJECTS

ONR supported previous research on "Magnetic Anomaly Detection by Remote Means" (N00014-13-1-0282) and is supporting ongoing follow-on research on "Stand-Off Detection of Magnetic Anomalies by Circular Polarized Laser Interactions" (N00014-15-1-2185). ONR is currently supporting "Stand Off Detection of Trace Species by Simultaneous Radar REMPI and Backwards Lasing" (N00014-15-1-2656).

PUBLICATIONS

1. A. Dogariu and R. B. Miles, “Three-photon femtosecond pumped backwards lasing in Argon,” *Opt. Express* **24**, A544-A552 (2016).
2. A. Dogariu, M. N. Shneider, and R. B. Miles, “Versatile Radar Measurement of the Electron Loss Rate in Air,” *Appl. Phys. Lett.* **103**, 224102 (2013).
3. A. Dogariu, J. B. Michael, and R. B. Miles, “Standoff stimulated emission in air,” *Proc. SPIE* **8366-20**, (2012).
4. R. B. Miles, A. Dogariu, and J. B. Michael, “Bringing Bombs to Light,” *IEEE Spectrum* **49**, 38 (2012).
5. A. Dogariu, J. B. Michael, M. O. Scully, and R. B. Miles, “High Gain Backward Lasing in Air,” *Science* **331**, 442 (2011).
6. J. Michael, M. R. Edwards, A. Dogariu, and R. B. Miles, “Femtosecond laser electronic excitation tagging for quantitative velocity imaging in air,” *Appl. Opt.* **50**, 5158 (2011).
7. A. Dogariu and R. B. Miles, “Detecting localized trace species using Radar REMPI,” *Appl. Opt.* **50**, A68 (2011).
8. J. Roslund, O. M. Shir, A. Dogariu, R. Miles, and H. Rabitz, “Control of nitromethane photoionization efficiency with shaped femtosecond pulses,” *J. Chem. Phys.* **134**, 154301 (2011).

Conference Presentations

9. A. Dogariu and R. Miles, Remote Backward-Propagating Lasing of Nitrogen and Oxygen in Air”, Conference on Lasers and Electro-Optics CLEO’2015, San Jose, CA (2015), invited.
10. R. B. Miles “The Use of Radar for the Characterization of Laser-Generated Plasmas and for Stand-Off Trace Gas Detection”, 17th International Symposium on Laser-Aided Plasma Diagnostics Hokkaido, Japan, September 27 – October 1, 2015
11. A. Dogariu, “Remote Trace Detection of Hazardous Substances using Nonlinear Optics,” Light, Energy and Environment Congress, Canberra, Australia (2014), invited.
12. R. B. Miles and A. Dogariu, “"Flow Imaging and Standoff Detection by Dissociation of Air Molecules," in Imaging and Applied Optics 2014, OSA Technical Digest, LW2D.1 (2014).
13. R.B. Miles and A. Dogariu “Diagnostics by Dissociation: FLEET, lasing in air, and trace detection of complex molecules by Radar REMPI”, 2015 AIAA Aviation Forum, Atlanta GA. (June 17, 2014) (invited)
14. Tat Loon Chng, Richard Miles, “Absolute concentration measurements of atomic oxygen in a flame using radar REMPI” (AIAA 2014-1360) 52nd Aerospace Sciences Meeting, 2014, 10.2514/6.2014-1360
15. A. Dogariu, R. B. Miles and “Backwards nitrogen double lasing in air for remote trace detection," in *Imaging and Applied Optics 2014*, OSA Technical Digest, LW2D.3 (2014).
16. A. Dogariu and R. B. Miles, "Lasing in Atmospheric Air: Similarities and Differences of Oxygen and Nitrogen," in *Frontiers in Optics 2013*, OSA Technical Digest, LTh2H.2 (2013).

17. R. B. Miles, “Nonlinear Processes in Air,” International Conference on Coherent and Nonlinear Optics (ICONO 2013) International Conference on Lasers, Applications, and Technologies (LAT 2013) 2013, June 18-22, 2013 Moscow, Russia
18. A. Dogariu and R. B. Miles, “Nitrogen Lasing in Air”, Conference on Lasers and Electro-Optics CLEO’2013, San Jose, CA (2013)
19. J. B. Michael; T. Chng; S. Zaidi; A. Dogariu; R. B. Miles, “ Species Concentration Measurements in a Pulsed Subcritical Microwave-Enhanced Flame”, AIAA-2012-0378, AIAA Aerospace Sciences Meeting, Nashville, TN, Jan 9-12, 2012
20. J. Michael, A. Dogariu, and R. Miles, “Strongly Correlated Atomic Oxygen Lasing In Air With Nanosecond Pumping,” AIAA 2012-3089 43rd AIAA Plasmadynamics and Lasers Conference, New Orleans, LA June 25-28, 2012.
21. A. Dogariu, J. B. Michael, A. V. Sokolov, M. O. Scully, and R. B. Miles, "Correlations and collisions in air laser emission for atmospheric remote sensing", Conference on Lasers and Electro-Optics CLEO’2012, San Jose, CA (2012).
22. A. Dogariu, J. Michael, and R. B. Miles, “Remote backwards emission in air via stimulated emission in atomic oxygen,” Laser Applications to Chemical, Security and Environmental Analysis (LACSEA), San Diego, CA (2012).
23. A. Dogariu, J. Michael, and R. Miles, “High gain atomic oxygen lasing in air,” 42nd AIAA Plasmadynamics and Lasers Conference, 2011-4001, Honolulu, HI (2011).
24. A. Dogariu, J. B. Michael, M. O. Scully, and R. B. Miles, "Remote backwards lasing in air", Conference on Lasers and Electro-Optics CLEO’2011, Baltimore, MD (2011).
25. A. Dogariu, M. Shneider, and R. Miles, “Measurement of Electron Loss Rates in Atmospheric Pressure Air by Radar REMPI,” 49th AIAA Aerospace Sciences Meeting, 2011-1324 Orlando, FL (2011).

Special Seminars

R. B. Miles, “Pumping Air: FLEET, Radar REMPI and Backward Lasing New Methods for Measuring Flow Properties and Contaminants in Air” Midwest Mechanics Seminar invited speaker

Tour A: University of Michigan, Michigan State University, Notre Dame, Northwestern University, University of Wisconsin Oct. 12-16, 2015

Tour B: Purdue, University of Illinois Urbana Champaign, Illinois Institute of Technology, Iowa State University, University of Minnesota Oct. 29-30, 2015

Students supported

1. James Michael (Ph.D. June 2012, currently Assistant Professor at Iowa State University, Ames Iowa.)
2. Sean McGuire (Ph.D completed August 2015, currently Post Doctoral associate at the Ecole Centrale, Paris)
3. Tat Loon Chng (will complete Ph.D. in October 2016)
4. Chris Limbach (Ph.D. completed in June 2015, Post Doctoral position at the Colorado State University)

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